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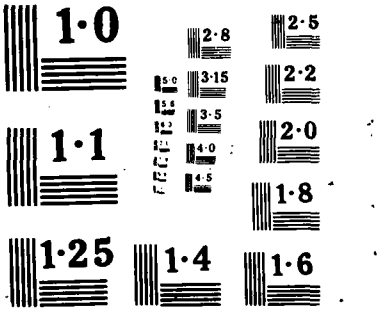
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TECHNICAL REPORT BRL-TR-2740

PHOTOCHEMICAL IGNITION STUDIES. II. OXYGEN-ATOM TWO-PHOTON RESONANCE EFFECTS

Brad E. Forch
Andrzej W. Miziolek

June 1986

US ARMY BALLISTIC RESEARCH LABORATORY
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Ultraviolet lasers have recently been used to activate and ignite reactive gas mixtures through multiphoton photochemical means. The observation of this new phenomenon opens up the possibility of using uv lasers as primary ignition or ignition augmentation sources for igniting propellants or their pyrolysis products. The first report in this series describes the ignition of hydrocarbon/air, N_2O mixtures by the excimer laser lines KrF (248 nm) and ArF (193 nm). In this report, a sharp wavelength dependence on the amount

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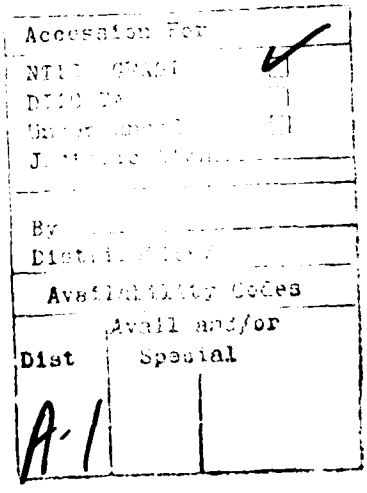
of incident laser energy necessary to ignite a premixed flow of H_2/O_2 at atmospheric pressure is described. This wavelength dependence exhibits a spectral profile similar to the two-photon fluorescence excitation curve for flame oxygen atoms and the respective peaks correspond to exactly the same wavelength near 225.6 nm. This similarity clearly indicates that oxygen atom production and subsequent excitation is an important step in the efficient (ca. 0.5 mJ) laser ignition of H_2/O_2 flows in this wavelength region. In addition, the dependence of the incident laser energy on the equivalence ratio reveals that the most efficient ignition occurs far into the fuel-lean region. This behavior further underscores the importance of the interaction between the uv laser and the oxidizer (O_2) component in the ignition of this reactive mixture.

TABLE OF CONTENTS



INTRODUCTION

Lasers have been used to ignite reactive mixtures for many years.[1,2,3] In most of this work, high peak-power lasers were employed and the ignition mechanism involved laser "spark" formation in the focal volume as a result of gas breakdown. The general applicability of this phenomenon to ignition studies, however, was found to have two serious limitations. One, since the process of gas breakdown has a sharp energy threshold, control of the amount of energy deposited in the focal volume is quite difficult. Two, because of this threshold effect, the laser produced spark is usually accompanied by a substantial blast wave which can be sufficiently intense to cause a transition into detonation (for detonable gas mixtures)[1,2] or to extinguish an already stabilized flame on a laboratory burner.[3]

The advent of commercially available pulsed uv lasers has recently enabled the ignition of reactive gas mixtures to be accomplished by a different mechanism that involves uv laser-induced photochemistry.[4,5] In fact, photochemical ignition had been demonstrated and studied some time ago through the utilization of the flash photolysis technique.[6] All of the photochemical work thus far has been carried out in the single-photon regime. Quite recently, however, we have performed a series of experiments in our laboratory that involved multiphoton photochemical ignition of reactive mixtures consisting of simple hydrocarbons and air or N_2O as oxidizers.[7] We reported therein, for example, that the ArF (193 nm) excimer laser could ignite a mixture of C_2H_2/ air with as little as 0.25 mJ of incident laser energy. This behavior is explained by the strong photochemical interaction between the focused ArF laser and the C_2H_2 molecules which produces many reactive intermediates, i.e., C_2H, C_2, CH, H and C radicals, and C^+ ions. We report here an extension of this initial work to a different reactive system, namely, H_2/O_2. In this work we have observed not only the photochemical interaction between the uv laser and the parent O_2 molecules to produce O atoms, but also a strong wavelength dependence for igniting H_2/O_2 which is related to oxygen atom two-photon excitation. To our knowledge, these results are the first example of such a strong wavelength dependence in the ignition properties of premixed gases.

II. EXPERIMENTAL

The experimental set-up is very similar to that previously reported,[8] and is shown in Figure 1. Tunable laser radiation in the 225.6 nm region was focused with a 50 mm focal length lens at a position 1-2 mm above the burner surface. The water-jacketed H_2/O_2 burner was fabricated from a stainless steel Swagelock 0.25 in. terminator fitting through which a 0.9 mm hole was drilled. Matheson (Model 620) flowmeters were calibrated by a GCA Precision Scientific wet test meter for H_2 and O_2 flows up to 2 LPM. This resulted in orifice linear flow velocities in the 10^3 cm/sec range. The incident laser energies were always measured just before the focusing lens with a Scientech (Model 38-0103) disc calorimeter-power/energy meter. The emission signals were detected, averaged, and processed as described in Reference 8. The excitation wavelength scans were performed manually, and each emission wavelength data point represented the average value for 512 laser shots.

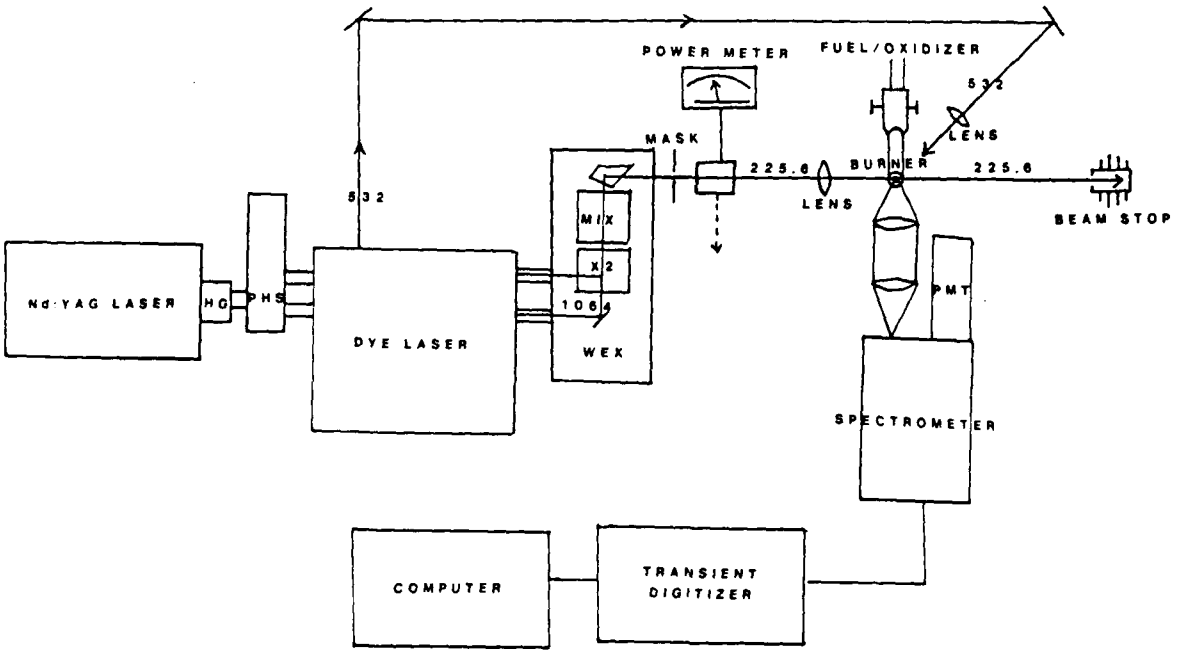


Figure 1. Experimental Schematic

III. RESULTS AND DISCUSSION

Figure 2 shows the dependence of the incident laser energy required to ignite a fuel lean, ϕ = 0.6, (Figure 2a) and stoichiometric (Figure 2b) flow of H_2/O_2 with the focal point fixed at the same distance above the burner surface. There are two noteworthy features that should be mentioned. One, the width of the fuel-lean wavelength dependence curve is greater than that found for the stoichiometric curve (each curve representing six or more independent measurements). Two, the minimum point in the incident laser energy is lower for the fuel-lean case than for the stoichiometric case. The minimum laser energy value depends on the distance between the laser focal point and the burner surface with the lowest value (ca. 0.3 mJ) recorded in the near limit of the laser tangent to the burner surface. Even in this limit, the relative laser energy difference between the minimum of the two equivalence ratios is maintained. The difference in the widths is not yet well-understood and is currently the subject of further investigations.

Figure 3 depicts the two-photon fluorescence excitation curve for oxygen atoms in a stoichiometric H_2/O_2 flame. The monitored emission at 777.5 nm is a result of the collisionally-induced energy transfer process which populates the upper state (^5P) of this transition.[8] A comparison of Figures 2 and 3 elicits two noteworthy comments. One, even though the general shapes of all three curves are similar, both Figures 2a and 2b are much broader than the excitation curve. Two, the peaks of all three curves corresponds to exactly the same wavelength. This strongly implies that the excitation of oxygen atoms is an important feature of the ignition mechanism.

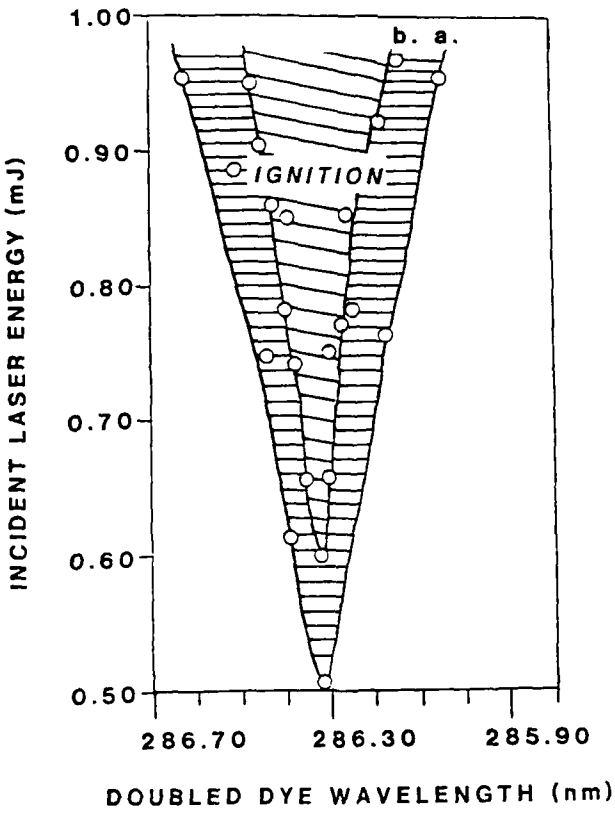


Figure 2. Incident Laser Energy Necessary to Ignite a Premixed Flow of H_2/O_2 Which is (a) Fuel-Lean (Equivalence Ratio = 0.6) and (b) Stoichiometric (Equivalence Ratio = 1) as a Function of Laser Wavelength in the 225.6 nm (Doubled Dye + 1.06 Micron) Region.

Figure 4 gives further evidence that the interaction between the uv laser and the oxidizer in the H_2/O_2 system is particularly important in the ignition process. The observed minimum in the incident laser energy occurs far into the fuel-lean region. This is in contrast to the very large body of previous spark ignition work which has determined that the minimum is generally quite close to an equivalence ratio value of one.[9] On the other hand, the very recent work on photochemical ignition of H_2/O_2 and H_2/air utilizing 157 and 193 nm radiation has also found the most efficient ignition in the fuel lean region due to the single-photon photolysis of the O_2 molecule.[4]

It should also be mentioned that all of our ignition experiments were performed on gas mixtures that flowed directly into laboratory air, whereas most of the previous measurements were made with gas mixtures irradiated in closed vessels. Advantages of our approach include the simplicity of apparatus and inherent safety since transitions into detonation are avoided. Disadvantages include the possibility of air entrainment (which was observed in our previous work),[7] as well as a more stringent laser energy requirement. This is due to the fact that additional mechanisms that do not affect the closed vessel work, such as a flow of cold gases as well as cooling by the

burner surfaces, may efficiently quench particularly weak ignition kernels. With respect to a possible air entrainment problem, we repeated the ignition experiments a number of times with the laser focal point translated horizontally ±1 mm from the center of the H_2/O_2 flow field. The results indicated very little change with respect to ignition behavior, and thus, the uncertainty in the equivalence ratios given in Figure 4 should be quite small.

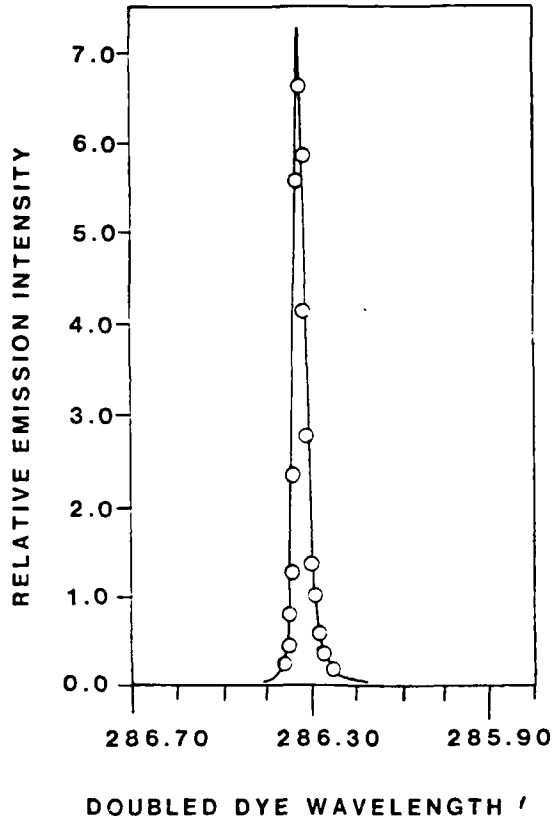


Figure 3. Two-Photon Fluorescence Excitation Spectrum for the J=2 Ground State Spin-Orbit Component of O-Atoms in a Stoichiometric H_2/O_2 Flame. The monitored emission is at 777.5 nm.

IV. CONCLUSION

Numerous questions still remain concerning the details of the mechanism involved in multiphoton photochemical ignition. We are, at present, involved in additional experiments in this area. Clearly, the results presented in this report have underscored the importance of the atomic excitation channel for the H_2/O_2 case with the implication that the chemistry of electronically excited O atoms, or the formation of O^+ ions due to the absorption of a third photon,[10] or both, may be key processes in the ignition mechanism. Also, the roles played by the possible photochemical production of metastable atoms or by the heat resulting from the photolysis step and/or from quenching, are not

clear at this time. More experimentation is necessary to characterize the relative importance of each of these factors before this phenomenon is amenable to modeling efforts. Still, the list of potential advantages that result from highly controlled energy deposition into a well-defined volume for the purpose of activating and/or enhancing reactive systems[7] can now be expanded to include the high efficiencies that are normally associated with resonance effects.

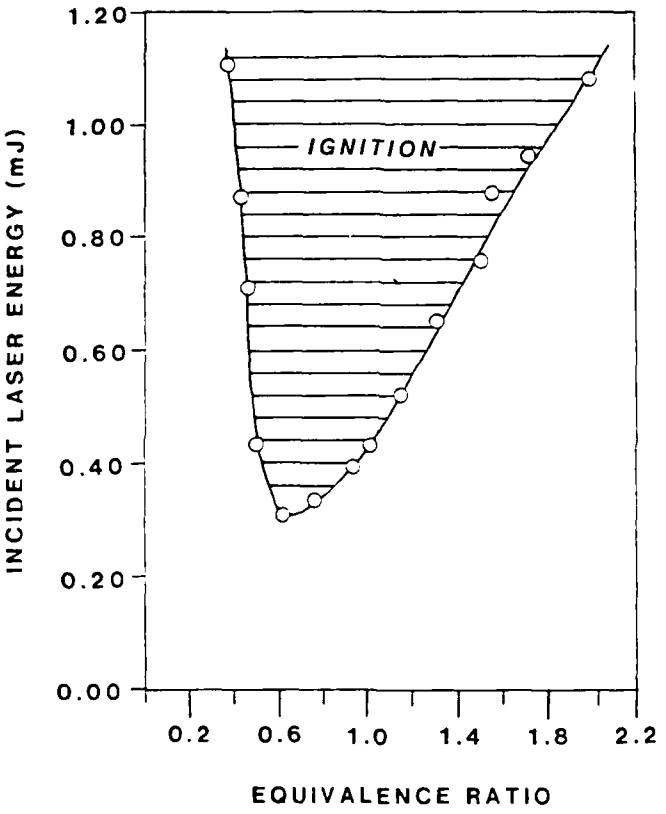


Figure 4. Dependence of the Incident Laser Energy Required to Ignite a H_2/O_2 Mixture as a Function of Equivalence Ratio. The laser wavelength is set at the peak for O-atom two-photon excitation.

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The authors thank Mr. Mark DeWilde for experimental advice on H_2/O_2 flows. (Mr. DeWilde is employed at the Ballistic Research Laboratory, Aberdeen Proving Ground, MD.) This research is supported in part by the U.S. Air Force Office of Scientific Research, Contract #85-0056.

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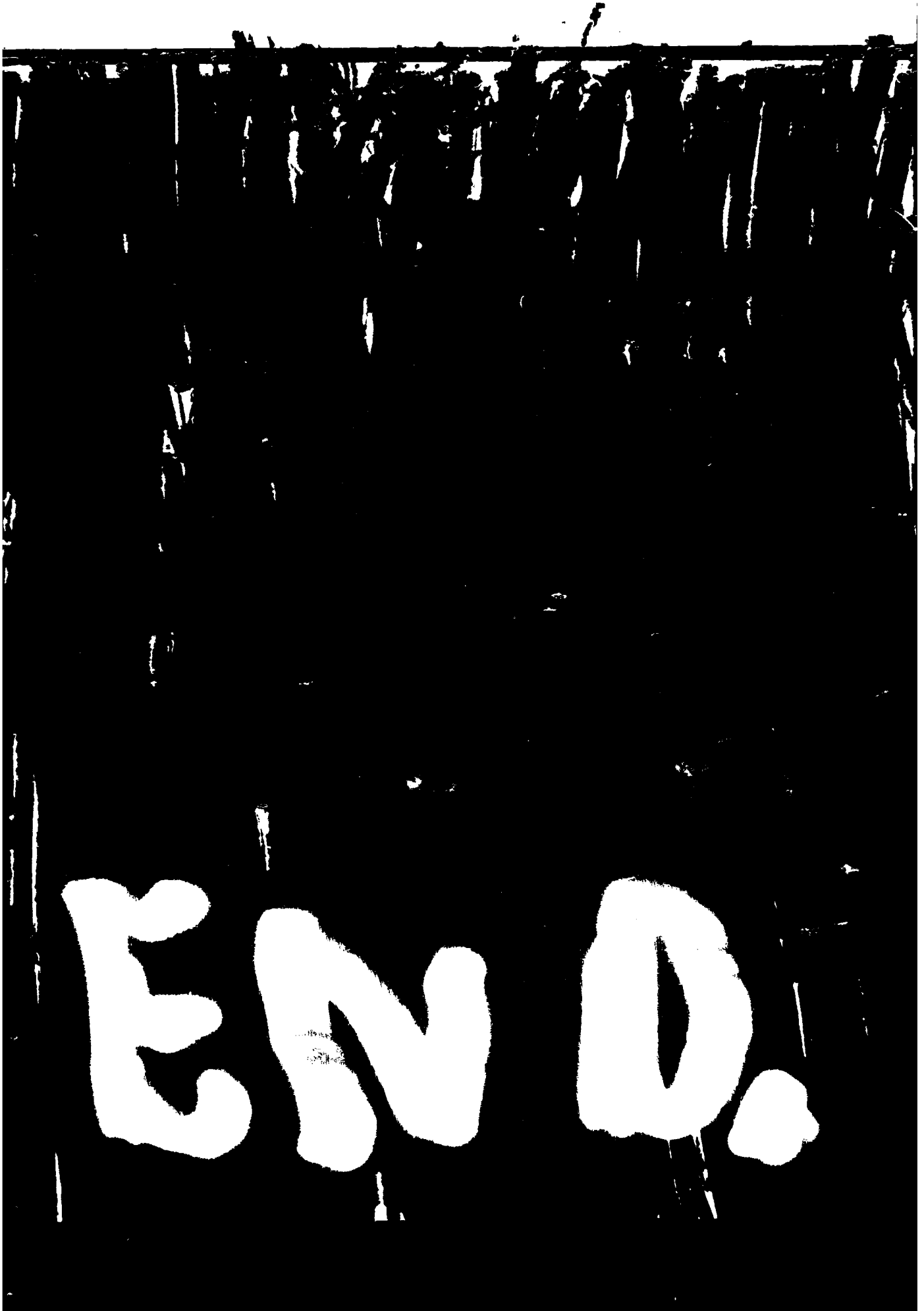
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